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Changes in PM_{10} concentration due to large-scale rainfall

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Abstract This paper is concerned with the estimation of the removal efficiency of PM_{10} by large-scale precipitation under no-wind conditions in a background (rural) and urban areas. The changes in PM_{10} concentrations before, during and after the presence of rainfall were studied from 2007 to 2013. The study was conducted in two different locations identified with regard to air quality. DAVIS weather stations were used to determine the meteorological conditions. The concentration of PM_{10} was calculated with the use of the gravimetric reference method. Two hundred and ninety-nine measurement series were carried out. A linear relationship was found between the intensity and duration of rainfall and the value of the removal coefficient (ΔC). It was proved that except light rains, for the near-to-ground troposphere, the effectiveness of the removal of PM_{10} (ΔC) did not assume different values at various locations for rainfall with the same intensity and duration. It was found that a temporary interaction of the effect of the purification by wet deposition was being minimised in areas characterised by low air quality. It was confirmed that intense rains resulted in the maintenance of higher values of air quality.

Keywords Urban area · Rural area · Particulate matter · Wet deposition · Frontal rainfall

This article is part of the Topical Collection on *DUST*

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Introduction

The influence of meteorological parameters on the scatter of pollutants around the sources of emission is beyond dispute. It is estimated that the volume and variability of pollution is largely affected by meteorological conditions, which can have an effect on both dilution and concentration of pollutants (Seinfeld and Pandis 1998; Wal and Janssen 2000; Turahoğlu et al. 2005). One of the principal processes that has an effect on the minimisation of the volume of solid particles is associated with their deposition.

There are two main forms of particle deposition by the troposphere: dry and wet. In the case of wet deposition, "rain scavenging is generally classified as rainout - particles serving as cloud condensation nuclei or undergoing capture by cloud water and as washout - the removal of below-cloud particles by raindrops as they fall" (Chate et al. 2003, p. 2477). In Poland, because of the specific structure of emitted particulate matter, dry deposition has a limited impact on atmospheric purification by suspended particles. On the other hand, below-cloud purification by rain is one of the major phenomena that controls the removal of atmospheric pollutants from the air. Hence, it is one of the most important processes that ensures that a balance is maintained between the source and outflow of aerosol particles (Chate et al. 2003). Wet below-cloud purification involves all the phenomena by means of which particles are removed from the air through a number of various types of precipitation: rain, snow, fog and ice. According to Bae et al. (2006), from the point of view of human health and the quality of the atmosphere, below-cloud scavenging seems to be more important than in-cloud scavenging, as particles of all types and sizes are deposed and transmitted to the ground-level zone in this process. This statement is supported by the fact that the particulate matter (PMs) that form an immediate hazard to human health are usually deposed as a result of below-cloud purification, and the principal mechanism involves the collision of solid particles with rain droplets (Kim et al. 2012).

Below-cloud scavenging of aerosol particles is usually described based on the concept of collisions between rain droplets and particulate matter. Nevertheless, the mechanisms of below-cloud purification, including the effect of inertia, Brownian diffusion, thermophoresis, diffusiophoresis and electro-scavenging, have been thoroughly recognised and described (Wang and Pruppacher 1980; Pruppacher and Klett 1997; Tinsley et al. 2001; Chate et al. 2003, 2011; Andronache 2004; Bae et al. 2009; Feng 2009; Santachiara et al. 2013).

The actual effects of washout of particles accumulated in the troposphere during episodes of precipitation are usually determined by means of the scavenging coefficient Λ (s^{-1}) (Radke et al. 1980), which is considered to be the most important parameter characterising below-cloud scavenging (SAI 2005; Environ 2008).

The literature contains much information about removal mechanisms and field experiment results connected with rainfall efficiency in scavenging processes (Laakso et al. 2003; Viana et al. 2003; Castro et al. 2010; Connan et al. 2013). The authors tend to focus on the effectiveness of the washout of solid particles of a specific size under specific rainfall types, and there is a lack of information about long-term field experiments on the effectiveness of the purification by large-scale precipitation of varying intensities in the absence of wind and direct insolation conditions. With minor exceptions (Laakso et al. 2003; Tai et al. 2010), experimental research is limited to continuous yet short-term observations (Davenport and Peter 1978; Schumann 1989; Maria and Russell 2005) or regards several instances over a small span of time and pertains to one type of precipitation (Jylha 2000; Chate and Pranesha 2004; Chate 2005; Zhang et al. 2012). With the exception of Schumann (1989), Maria and Russell (2005) and Zhang et al. (2012), authors do not consider the case of coarse particle removal, which constitutes the standard indicator of air quality. In addition, authors here do not consider the variability of aerosol concentration over the several hours after the termination of the rainfall.

This paper reports the results of a study involving the assessment of the effectiveness of PM_{10} removal by large-scale precipitation, which occurs in windless conditions in two areas with different aerosanitary parameters. The principal aim of this paper involves the analysis of the variability of removing coarse aerosols relative to the duration and intensity of precipitation and location in which the process of wet deposition occurs. The auxiliary objectives include an attempt to develop a simple linear model to describe the effectiveness of PM_{10} removal by large-scale precipitation based on experimental results and an attempt to determine the effect of the location of the process and parameters that characterise frontal precipitation on the effectiveness of recurrently enriching the atmosphere with PM_{10} after the termination of the process of precipitation. This paper includes five hypotheses:

- The effectiveness of removing PM_{10} is not relative to the season associated with the occurrence of large-scale precipitation (I).
- There are no differences in the effectiveness of PM_{10} removal depending on the location of frontal precipitation with the same intensity and period of duration (II).
- Location does not affect the rate of the increase in the concentration of particulate matter after precipitation with the same intensity and period of duration (III).
- Season in which the process occurs does not have any effect on the rate of the increase in PM_{10} after frontal precipitation (IV).
- The intensity of 3-h large-scale precipitation does not affect the rate of variability in the concentration of PM_{10} after the termination of the rainfall (V).

Materials and methods

Measurement site description and experiment principle

The study was conducted simultaneously in urban and background (rural) areas over 7 years (2007–2013). The registration of meteorological parameters and concentration of PM_{10} in the urban area were realised in the provincial town of Opole (Poland, 50° 41′ 13″ N; 17° 56′ 43″ E, 122,000 inhabitants). The measurement point was situated in the north-east part of the town, near low residential housing and commercial development, and was close to a major road into the town with moderate traffic. The meteorological data representative of a non-urban area and concentration of PM_{10} was measured in an undeveloped area, i.e. near a village (Kotórz Mały, Poland, 50° 43′ 37″ N; 18° 03′ 22″ E; 1025 inhabitants). The second measurement point was located in an open, yet shielded, meadow area protected by the surrounding wood—9 km north-east from the border of Opole and 2 km from the nearest compact rural building development. The 5-year assessment of air quality published by Opole Voivodeship Inspectorate for Environmental Protection (OVIEP), which involves the analysis of air pollution with particulate matter (PM_{10} and $PM_{2.5}$), indicated that Opole is classified as class 3b[1] with regard to the quality criteria for the protection of human health (OVIEP 2014a). This urban area is characterised by considerably

[1] Class 3b—an area in which the level of concentration of pollution is higher than the upper assessment threshold established for this type of substance in the air; this level invokes a need to conduct intensive and continuous registration of data in measuring stations in the areas exceeding the admissible levels

higher mean annual values of the concentration of PM_{10} compared to the distant and partly isolated rural area. For instance, according to OVIEP data (OVIEP 2014b), the mean annual concentration of PM_{10} in Opole was equal to 33.3 μg m^{-3} in 2013, which is 83.2 % of the admissible level. This result is confirmed by the mean annual concentration of PM_{10} measured in the present study (36.4 μg m^{-3}), which was recorded at the observation point. Concurrently, the mean annual concentration of PM_{10} measured in the rural area was about 40 % lower.

The measurement campaign involved the observation of the PM_{10} concentration resulting from the occurrence of large-scale (frontal) precipitation with different intensities and durations. To minimise the additional impact of mass transport on aerosol concentration levels, all observations were conducted under no-wind conditions. In order to further verify the results of the research, the analysis involved only the cases of wet deposition occurring simultaneously at both measurement points. The detailed analysis involved episodes of large-scale wet deposition that occurred simultaneously and were characterised by the nearly identical intensity of precipitation R (mm h^{-1}). The time interval of the particular aspirations was constant and equal to 30 min. Both short-term (0.5 h) and long-term ($1 < 6$ h) instances of precipitation were considered in the analysis. For the purposes of the presentation and analysis of the results, precipitation was classified into the following categories: light (R <0.4 mm h^{-1}), intermediate (R 0.4–3.9 mm h^{-1}) and heavy (R >4.0 mm h^{-1}). In addition, 40 separate observations were conducted (each lasting for 6 h with a time interval of 30 min) with regard to the variability of the PM_{10} concentration directly after the termination of the rainfall.

Meteorological data and PM_{10} sampling procedure

To determine the meteorological conditions, portable weather stations (DAVIS®) were used, which are widely used for the registration of weather conditions in field measurements (Castro et al. 2010). These weather stations were installed 10 (in the city) and 12 m (in the background area) from the PM aspirators. At both measurement points, the sensors, which determined relative humidity (RH), temperature (T), atmospheric pressure (Ap), wind speed (Ws), wind direction (Wd) and rainfall intensity (R), were installed at a height of 2 m above the ground. The standard measurement uncertainty was equal to RH 0.5 %, T 0.5 °C, Ap 0.06 hPa, Ws 0.06 m s^{-1} and Wd 1°, respectively. The stations were equipped with automatic sequential samplers for precipitation measurements. Due to the capacity of the overflow bucket (2 ml), the stations were insusceptible to precipitation with very low intensity. Consequently, the results did not account for precipitation with an intensity below 0.2 mm h^{-1}.

The procedure by which the measurement of the concentration of PM_{10} was performed was in conformity with the European standard BS EN 12341:1999. The reference method, which is often relied on (Chate et al. 2003; Connan et al. 2013), was also applied in this case. At both measurement sites, the aspiration of the PM_{10} in the air was carried out with a MicroPNS HVS16 (UMWELTTECHNIK MCZ GmbH®) sequential dust sampler. Like the sensors in the weather stations, the aspiration headers were installed 2 m above ground level. The flow rate was 68 m^3 h^{-1}. The PM separators applied were Whatman GF/A fibreglass air filters with a diameter of 150 mm. Prior to and after aspiration, the filters were seasoned over 24 h in conditions of constant temperature and humidity and, subsequently, their weight was determined by a differential scale (RADWAG XA 52/2X). The aspiration at a constant time interval of 0.5 h was conducted directly before, during and after the occurrence of precipitation. The expanded concentration measurement of uncertainty (U) did not exceed 3.2 %. The time interval guaranteed the PM collection to a degree that was sufficient to determine the mass of the captured particulate matter, even in conditions when its concentration in the air was low (EC Working Group 2000). The initial testing ($n = 25$, time interval of registration—10 s, time of a single registration—1800 s) using a DustTrak 8520 Aerosol Monitor—TSI® was conducted in variable weather conditions; however, with the exception of rain, it did not yield considerable differences in the results of PM_{10} concentration over 10 and 1800 s in the investigated background area.

Procedure for determining the removal efficiency coefficient ΔC

As was mentioned before, the scavenging coefficient Λ (s^{-1}) is customarily used to explain the effectiveness of the self-purification of the atmosphere. However, this term can seem complicated to an everyday reader, and it does not explain directly the scale of the process, since "the man in the street" could have problems interpreting the results of Λ reported here. For convenient presentation, the effectiveness of PM_{10} scavenging by precipitation can also be shown as a simple relationship of percentage change (ΔC) in the concentrations before (C_0) and after (C_t) episodes of rain (1):

$$\Delta C = \frac{C_t - C_0}{C_0} \times 100\% \quad (1)$$

The proposed solution has a primarily practical character and constitutes an attempt to offer a way of approaching the effect of scavenging of particulate matter suspended in the ground-level zone.

Statistical analysis of the results of meteorological parameters and variability of the concentration of PM_{10} for verification of the research hypotheses was undertaken using the STATISTICA 12® programme.

Results and discussion

Short characteristics of meteorological parameters

Table 1 contains a summary of data regarding selected meteorological parameters and the precipitation registered during the 7-year experiment. Table 1 involves only the records for those days when liquid, large-scale precipitation occurred at both observation points. None of the recorded cases were found to correspond to a normal distribution of data, as indicated by the Shapiro-Wilk test. By considering the results regarding temperature, rainfall intensity and relative humidity with regard to the investigated location, one can note that the results are similar. Both observations points were very close and the processes of wet deposition have an identical origin. During large-scale rain in the cold season, the average air temperature in the urban area was around 2 °C higher in comparison to the rural area. A slightly smaller difference was also noted for the warm season. The origin, as well as the large-surface nature of the precipitation, resulted in a lack of statistically valid differences in its intensity in both areas during the same period of occurrence. In addition, no statistically valid differences were noted with regard to the value of relative air humidity. At the same time, the initial analysis did not yield any correlations between the effectiveness of PM_{10} scavenging from the near-to-ground troposphere and the meteorological parameters defining the conditions during the observations.

The key parameter used for determination of the effectiveness of particulate matter removal from the troposphere is the intensity of precipitation R. The analysis of this parameter indicates that, for both locations, the dominant form of wet deposition was rainfall with an intermediate intensity (around 45 %). The rarest form was accompanied by rainfall with a high intensity (around 23 %).

Effectiveness of PM_{10} scavenging and regression models

In order to simplify the understanding of the effect of solid particle scavenging as a result of the occurrence of large-scale precipitation in windless conditions, the results of the experiment had to be presented with the use of a coefficient that describes the effectiveness of the removal of particulate matter (ΔC (%)) with a diameter smaller than 10 μm. The collective results, separately for the background and urban areas, are presented in Fig. 1. The interpretation of the graphical results indicates that the effectiveness of removal increases along with the increase in the intensity and duration of frontal precipitation regardless of the period and location of its occurrence. The effectiveness of removing atmospheric aerosol is most often described in terms of a median derived based on measurements. The detailed comparative analysis of the medians indicates that, for constant rainfall intensities and their variable durations, regardless of the area of occurrence, nearly

Table 1 Meteorological parameters characterising the conditions during the observations

Precipitation period and location	Number of rain episodes[a]	Descriptive statistics	T (°C)	RH (%)	Ws (m s^{-1})
Urban—cold season (Oct–Mar)	78	Avg	9.8	0.83	0.13
	L (24)	Med	8.9	0.86	0.10
	M (36)	Min	−1.0	0.73	0.00
	H (18)	Max	14.4	0.99	0.30
Urban—warm season (Apr–Sep)	72	Avg	17.6	0.85	0.11
	L (24)	Med	15.4	0.88	0.08
	M (30)	Min	13.1	0.69	0.00
	H (18)	Max	28.6	0.94	0.20
Rural—cold season (Oct–Mar)	59	Avg	7.6	0.85	0.10
	L (19)	Med	7.7	0.89	0.10
	M (26)	Min	−1.5	0.66	0.00
	H (14)	Max	11.2	0.94	0.20
Rural—warm season (Apr–Sep)	50	Avg	15.9	0.79	0.14
	L (15)	Med	14.3	0.81	0.11
	M (24)	Min	9.1	0.69	0.00
	H (11)	Max	27.9	0.95	0.30

[a] In the brackets are no. of episodes for light (L), 0.2–0.4 mm h^{-1}; moderate (M), 0.5–3.9 mm h^{-1}; and heavy (H), >4 mm h^{-1} rains, respectively

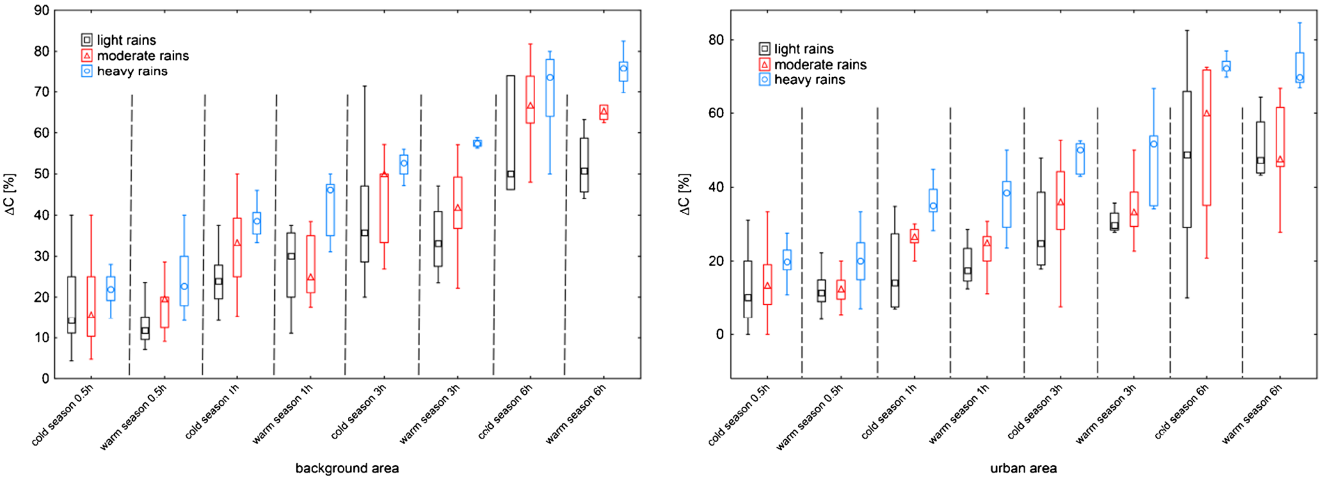

Fig. 1 Effectiveness of PM_{10} scavenging in the function of the duration and intensity of large-scale precipitation in the examined locations

constant values of the ratios (ΔC_{6h} to $\Delta C_{0.5h}$ (3.6 and 3.8), ΔC_{3h} to $\Delta C_{0.5h}$ (2.4 and 2.5), ΔC_{1h} to $\Delta C_{0.5h}$ (1.7 and 1.8)) are observed for the cold and warm seasons, respectively. A relevant dependence was also observed in relation to the rainfall intensity, an increase in which, for a constant duration of wet deposition, takes on the form of a positive correlation. For example, the analysis indicated that, during the cold season, the effectiveness of scavenging PM_{10} in the rural area from the air was about 26 % higher for the case of heavy rains than for light ones. In the warm season, this relation was around 50 %. In the urban areas, these values were higher, and they were equal to 65 and 56 %, respectively. Nevertheless, for the case of large-scale rains, a considerable scatter of the value of ΔC was observed for the particular ranges of rainfall intensity. This condition is particularly valid in the case of precipitation with light and intermediate intensities. Concurrently, the coefficient of variability (C_v) slightly decreased along with an increase in intensity and duration of precipitation. The reason for this condition can be attributed to the variable structure of wet deposition (Chate et al. 2003; Zhang et al. 2004). In the examined range of intensity R, it is possible to identify instances of drizzle (small, dense droplets) as well as the deposition of coarser drops with larger diameter, which affects the effectiveness of atmospheric purification. Obviously, the results gathered in variable thermal conditions of the environment tend to confirm a statement that the effectiveness of particulate matter removal is affected by a number of factors, including turbulence in the boundary layer, chemical processes in the wet phase, acceleration/reduction in convective motion and the emission of pollutants from remote areas (although, in this case, it is limited due to the lack of horizontal air mass motion) (Laakso et al. 2003). In addition, for the case of light rainfall with identical intensities and durations, the comparison of the medians of the calculated removal coefficient ΔC indicates that the effectiveness of the removal of suspended particulate matter is slightly higher in the case of non-urban areas. These divergences are smaller for intermediate and light rains.

The basic statistical analysis using Spearman correlation confirmed the results of the raw data and indicated a considerable degree of relation (around 0.91) between the effectiveness of the removal of ΔC and the duration and intensity of precipitation. The calculated determination coefficient could justify the variability in the value of ΔC, which occurs due to the varying intensity and duration of precipitation. The analysis of the results gathered for the same values of R suggests that, with regard to short-term rainfall (0.5 h), the independent variables (t and R) explain the variability of PM_{10} concentration to around 94 % in the cold season (C) and around 86 % in the warm season (W). In the case of the urban area, this value is equal to 86 and 83 %, respectively. The determination coefficient r^2 derived for precipitation with a duration of 1 h was calculated at the level of about 76 % (C) and 79 % (W) for the urban area and around 85 % (C) and 79 % (W) for the rural environment. For the case of continuous precipitation lasting for 3 h, the value of r^2 was found to be equal to 86 % (C) and 83 % (W) in the rural area and 88 % (C) and 85 % (W) in the town. Continuous precipitation with high intensity and duration of 6 h can justify the variability in the concentration of particulate matter at around 81 % (C) and 85 % (W) for the undeveloped area and at 86 % (C) and 77 % (W) for the urban area. These differences could stem from non-uniform, local emission at the two measurement points (also occurring during the process of wet deposition), which is expressed by higher values of the variability coefficient ΔC (by about 15 %) derived in the urban area during the episodes of precipitation with the same intensity. The graphical interpretation of the results reveals inconsiderable differences in the value of ΔC calculated in the examined seasons at a given location for the specific duration and intensity of precipitation. Nevertheless, the conducted statistical analysis using the Mann-Whitney U test, for the relevance level of

$\alpha = 0.05$, indicates that there were no statistically relevant differences in the effectiveness of PM_{10} removal in the cold and warm seasons (calculated separately in the urban and rural areas). The lowest calculated value of probability (*p* value) was equal to 0.08 for heavy rains in the rural area.

Table 2 summarises the results of the Mann-Whitney *U* test, which verifies the hypothesis that there are no considerable differences in the effectiveness of PM_{10} removal relative to the location of the precipitation with the same intensity and duration.

The comparison of areas with different aerosanitary characteristics (urban vs. background) indicated that, for short-term rainfall (0.5 h), there were considerable differences in the effectiveness of particulate matter removal in the cold season for the case of precipitation with low intensity and during the warm season for precipitation with intermediate intensity. The result of the test also indicated the existence of statistically relevant differences for the case of continuous precipitation (6 h) in the warm season. For the remaining cases, no significant differences were noted. The results were reliable due to the involvement of a considerable volume of data. The differences noted during the cold season could be attributed to the high emission of particulate matter from stationary and mobile anthropogenic sources, which were present in the urban area. The large volume of aerosol emissions introduced into the air tended to effectively reduce the value of the scavenging coefficient, which described the removal of particulate matter. However, it was difficult to justify the noted divergences in the warm season. The rainfall with the intermediate intensity tended to have considerable effectiveness when purifying the atmosphere of particulate matter. The results of the test indicated that, during the continuous rains, the *p* value was on the border of the level of relevance. The low value of the probability test for short-term precipitation accompanied by considerably smaller values of ΔC for the urban area is noteworthy; however, it finds confirmation in the ten independent observations. On the other hand, eight of nine observations were made in May, which is characterised by intense pollen shedding from the chestnut trees located near the measurement point beside the existence of standard anthropogenic emission. The more intense natural emission could lead to a similar influence on the results as anthropogenic emission in the cold months.

It is believed that the effectiveness of particulate matter removal is relative to the duration and intensity of precipitation (González and Aristizábal 2012). In general, one could assume that, for the same location, intensity and duration of wet deposition, the effectiveness of PM_{10} removal is not affected by the season in which the process occurs. The considerable values of the linear correlation coefficient (Spearman), which accounts for the relation between the decrease in the particulate matter concentration in the air and duration and intensity of precipitation, could lead to the conclusion that the two independent variables (*R* and *t*) could be accounted for as prediction parameters in the modelling of the investigated relations. The above statement provided an attempt to find a simple model that can describe the effectiveness of PM_{10} removal in the areas with different aerosanitary characteristics. A classical analysis was undertaken, which involved the most basic foundations, stating that:

- The model assumes the stability in the function established between the examined phenomena.
- The model is linear in respect to the parameters of the expression (2) (Mark and Goldberg 1998):

$$Y = \beta_1 \times X_1 + \beta_2 \times X_2 + \beta_0 + \xi \quad (2)$$

where β_1, β_2 and β_0 are the structural parameters of the model, and the random component is a random variable with a normal distribution $N\,(0,\sigma^2)$.

The analysis of the results for large-scale precipitation, identified separately in the rural and urban areas, without distinction between the seasons despite the stability of the function between the examined phenomena, has indicated that it is impossible to identify a regression model. The analysis of the residuals indicated that the condition of the normality of the distribution was fulfilled. The probability of an error of the first order calculated by means of the Shapiro-Wilk test (for $\alpha = 0.05$) was equal to 0.03 and 0.04 for the town and village, respectively. It was possible to develop regression models only for the cold season for the case of non-urban area and for the warm season for the urban area.

The results of the analysis for large-scale precipitation lead one to conclude that the linear regression model accounting for independent variables *R* (mm h^{-1}) and *t* (h) can explain around 91 % of the variability in ΔC. The mean discrepancy between the actual values of the dependent variable and the values predicted by the model was equal to 5.5 (%). The high value of statistical *F* (above 1×10^2) and the level of probability corresponding to *p* ($p < 10^{-4}$) confirm the statistical

Table 2 The results of Mann-Whitney test

Observation (h)	Light rains		Moderate rains		Heavy rains	
	Cold	Warm	Cold	Warm	Cold	Warm
0.5	0.042	*0.186*	*0.189*	0.002	*0.167*	*0.269*
1	*0.191*	*0.221*	*0.121*	*0.363*	*0.210*	*0.340*
3	*0.202*	*0.327*	*0.091*	*0.083*	*0.144*	*0.201*
6	*0.723*	*0.713*	*0.417*	0.048	*0.748*	*0.631*

p value for two locations (urban-rural) in cold and warm periods of the year for different intensities of precipitation. Significance level $\alpha > 0.05$. Italic values show realisation of condition of Mann-Whitney test relevance of the linear model. The value of statistical t (almost 14), used to assess the relevance of the coefficient β_1 and its corresponding probability level $p < 10^{-5}$, confirms that this parameter was considerably different from zero. For the case of assessing the level of relevance of coefficient β_2 ($t = 7$; p value $= 0.003$), one can state as well that the value of this parameter was considerably different from zero. The normality of the residuals' distribution was verified based on the Shapiro-Wilk test. The calculated test probability was equal to 0.61, which made it impossible to reject the hypothesis regarding the normality of the distribution of the random component. The application of this model indicated that an increase in the duration of the precipitation instance by 1 h resulted in a decrease in the efficiency of removal by around 7.7 %, while an increase in the efficiency of large-scale precipitation during the cold season by 1 (mm h^{-1}) led to an increase in the efficiency of removal by 1.1 % (while the values of the remaining independent variables remained the same, in accordance with the ceteris paribus principle). The result gained in this manner confirmed the normality of the residuals' distribution. The resulting linear correlation between these parameters confirmed the normality of the random component.

The results of the analysis for large-scale precipitation occurring in the urban area and warm season showed that the model of linear regression accounting for independent variables R (mm h^{-1}) and t (h) offers justification to 92 % of the variability in ΔC. The mean difference between the actual values of the dependent variable and values predicted by the model was equal to 5.3 (%). The high value of statistical F (over 1.14×10^2) and the value of the probability corresponding to p ($p < 10^{-4}$) confirm the statistical relevance of the linear model. The value of statistical t (13.5), applied for assessing the relevance of coefficient β_1, and the corresponding value of probability ($p < 10^{-5}$) confirm that this parameter was considerably different from zero. For the case of the assessment of coefficient β_2 ($t = 6.6$; $p = 0.0004$), this parameter was different from zero. In the same manner, the extension of the duration of precipitation by 1 h resulted in an increase in the effectiveness of removal by 7.4 %, while an increase in the effectiveness of large-scale precipitation during the cold season by 1 (mm h^{-1}) resulted in an increase in the effectiveness of removal by 2.3 % (while the values of the remaining independent variables remained the same, in accordance with the ceteris paribus principle). The probability of the occurrence of the error of the first order (for $\alpha = 0.05$) was equal to 0.59.

The results of the regression analysis:

– For large-scale precipitation in the cold season for the non-urban area ($_{\text{C-N}}$):

$$\Delta C_{\text{C-N}} = \beta_1 t + \beta_2 R + \beta_0 \tag{3}$$

$\beta_1 = 7.8$, $\beta_2 = 1.1$, $SE_{\beta 1} = 0.56$, $SE_{\beta 2} = 0.36$, $\beta_0 = 16.35$, $SE_{\beta 0} = 2.4$, $r^2 = 0.91$, estimation error $= 5.47$, $t_1 = 13.74$ for $p < 0.0000$ and $t_2 = 7$ for $p < 0.003$.

– For large-scale precipitation in the warm season for the urban area ($_{\text{W-U}}$):

$$\Delta C_{\text{W-U}} = \beta_1 t + \beta_2 R + \beta_0 \tag{4}$$

$\beta_1 = 7.4$, $\beta_2 = 2.3$, $SE_{\beta 1} = 0.54$, $SE_{\beta 2} = 0.35$, $\beta_0 = 6.93$, $SE_{\beta 0} = 2.2$, $r^2 = 0.92$, estimation error $= 5.26$, $t_1 = 13.59$ for $p < 0.0000$ and $t_2 = 6.6$ for $p < 0.0004$.

Considering the large number of measurements and the results of the statistical analysis, it can be assumed that, with regard to the removal of coarse particulate matter, the dependence gained in the form of simple correlation can justify over 90 % of the variability in the effectiveness of the removal. Such a high ratio can lead to the conclusion that these relations are not only a matter of chance but also occur in the general population.

The growth rate of PM_{10} concentration after rainfall episodes

In the papers concerned with the efficiency of PM_{10} removal as a result of the effect of wet deposition, the authors focus on the process itself and its effects during the duration of the precipitation. There is a lack of information regarding the aerosanitary conditions that change after the termination of the process. In order to verify the effectiveness of atmospheric enrichment as a result of PM_{10}, a complementary series of measurements was undertaken with regard to particulate matter concentration after the termination of the precipitation. Figure 2 presents selected curves with the growth rate of PM_{10} concentration, which is representative of all types of precipitation in terms of intensity, period of occurrence and location.

For the case of the conditions following light precipitation ($R = 0.4$ mm h^{-1}), considerable differences can be observed between the rate of atmosphere enriching with PM_{10} in the urban and non-urban areas. During the cold season, the C_A/C_0 coefficient in the urban area was almost two times higher than that in the non-urban area. The measurements were realised at the ambient temperature close to 0 °C, which accompanied the intensive exploitation of the urban sources of point emission (i.e. ones associated with the individual supply with thermal power). A smaller disproportion was revealed by the results measured in the warm season (with a difference of around 30 %). With regard to the variations in PM_{10} concentrations that precede observations in the case of intermediate precipitation, the rate of growth in the PM_{10} concentration was higher in the urban area, which was characterised by the considerable

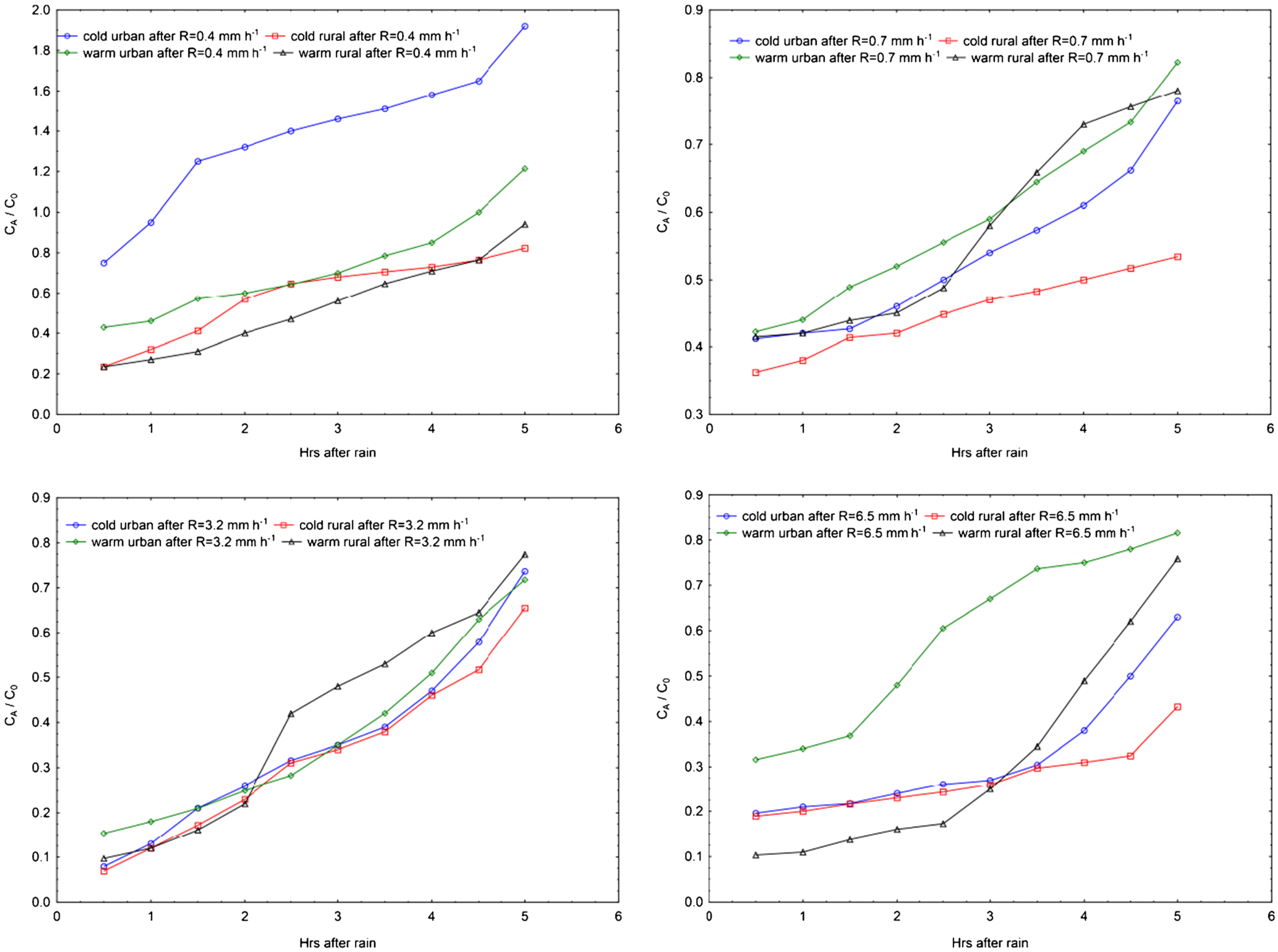

Fig. 2 The rate of PM_{10} concentration increase after selected rainfall episodes

number of anthropogenic sources. The difference was equal to as much as 13 % in the cold season, while this value was only 5 % during the warm season. In the town, for the case of the condition following the process of wet deposition with a high intensity, the value of the C_A/C_0 coefficient assumed higher values after the same duration of precipitation. While the rate of the change in the cold season was balanced (considerable difference is noted only after 3.5 h), the results for the warm season seem to be puzzling. Unfortunately, the results are not representative, due to road surface repair work in the direct vicinity of the measurement point.

Figure 3 presents collective data with a summary of changes in the concentration of particulate matter after 3 h rainfall episodes. The results are presented as the ratio of the concentration after precipitation (C_A) to the concentration directly before an episode of wet deposition (C_0). The results present the concentrations calculated up to 6 h after the termination of the precipitation. The registrations of PM_{10} concentration were performed at intervals of 0.5 h.

The analysis of raw data indicated that for the case of the remote area, the rate of the growth of the particulate concentration in the warm season is to a large degree dependent on the emission from the background (surrounding meadow and trees), as a small increase is noted during the first 2–3 h after the termination of precipitation and abruptly after the surfaces are dried after rain stops. In the case of the cold season, the rate of change in the PM_{10} concentration in the undeveloped area can be treated as uniform. A similar conclusion can be derived based on the results gained from the urban area in both cold and warm seasons.

Generally, it can be stated that, immediately after precipitation stopped, the coefficient C_A/C_0 was much higher after light rain than after heavy. On the other hand, the difference between C_A/C_0 for light and heavy rain slowly but consistently increased with time. The rate of change in the PM_{10} concentration as a local character was directly relative to all phenomena, both of natural and anthropogenic origin, which affects the level of particulate matter suspended in the near-to-ground troposphere. The results of the Mann-Whitney U test verified the hypothesis regarding the lack of differences in the growth rate in the concentration of particulate matter after precipitation with a constant

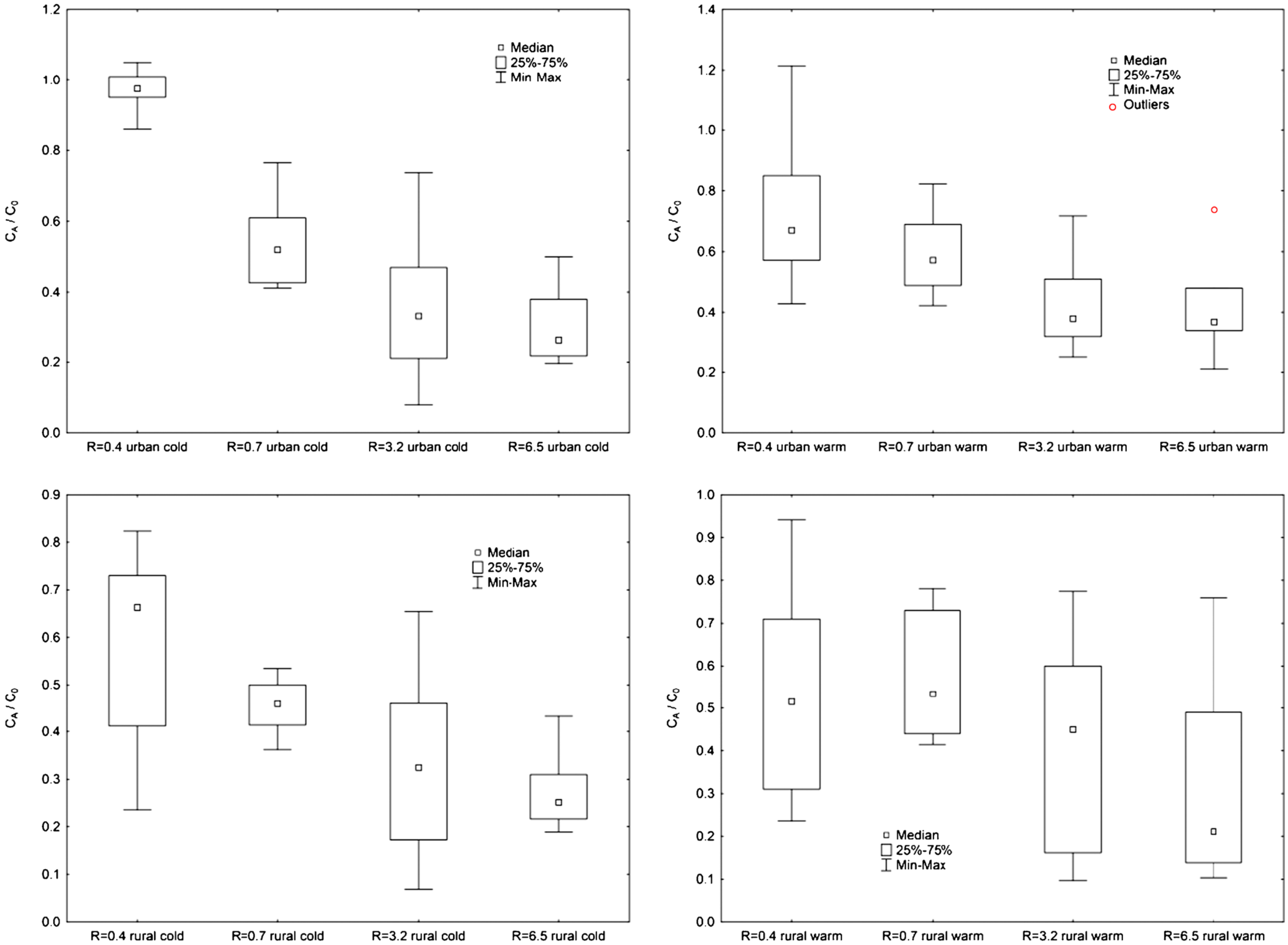

Fig. 3 Collective data regarding the rate of PM_{10} concentration increase after termination of precipitation with various intensities (*R*)

intensity and duration ($\alpha = 0.05$) performed in the two analysed locations. The results also indicated that statistically relevant differences could be noted only with regard to the conditions occurring after light precipitation ($p = 0.012$ for the cold season and 0.04 for the warm season). Following intermediate and heavy precipitation (with the exception of one abovementioned occurrence from the warm season), no statistically valid differences in the coefficient of the growth of PM_{10} concentration were registered. The verification of the hypothesis regarding the

Table 3 Summary results of Kruskal-Wallis test (nonparametric ANOVA)

Urban	Cold season			Rural
After 3 h rain with:	$R = 0.4$ mm h^{-1}	$R = 0.7$ mm h^{-1}	$R = 3.2$ mm h^{-1}	$R = 6.5$ mm h^{-1}
$R = 0.4$ mm h^{-1}		***0.912***	*0.029*	*0.001*
$R = 0.7$ mm h^{-1}	**0.075**		***0.342***	*0.031*
$R = 3.2$ mm h^{-1}	<0.001	**0.442**		***0.986***
$R = 6.5$ mm h^{-1}	<0.001	**0.202**		
Urban	Warm season			Rural
After 3 h rain with:	$R = 0.4$ mm h^{-1}	$R = 0.7$ mm h^{-1}	$R = 3.2$ mm h^{-1}	$R = 6.5$ mm h^{-1}
$R = 0.4$ mm h^{-1}		***0.943***	***0.899***	***0.334***
$R = 0.7$ mm h^{-1}	**0.923**		***0.701***	***0.100***
$R = 3.2$ mm h^{-1}	0.006	**0.159**		***0.431***
$R = 6.5$ mm h^{-1}	**0.853**	**0.723**		

p value for the growth rate of PM_{10} concentration after rainfall episodes with different intensity *R* (mm h^{-1}). Italic font indicates data for rural area. Bold values show realisation of condition of Kruskal-Wallis test lack of differences in the rate of the increase in aerosol concentration between the warm and cold seasons, performed with the same test, confirms this hypothesis with regard to non-urban areas for all circumstances following precipitation with intensities >0.4 mm h^{-1}. On the other hand, this hypothesis cannot be confirmed for urban areas for the rate of variations in the particulate concentration of <0.7 mm h^{-1}.

Table 3 contains detailed results of the Kruskal-Wallis test, which indicated the rate of growth in PM_{10} concentration following a 3-h occurrence of precipitation with various intensities performed in the same location. The basic results of the test indicated that the differences in the rate of particulate concentration growth did not indicate statistically relevant differences only in the conditions after precipitation in the rural area in the warm season ($p = 0.073$). In the remaining cases, the total value of the test probability did not exceed the level of relevance ($\alpha = 0.05$). The above indicated that the intensity of precipitation had a considerable impact on aerosanitary conditions in the urban area after the process of wet deposition.

Conclusions

The coefficient of particulate matter removal ΔC by precipitation can offer an alternative for the known solution used for the assessment of wet deposition as the process of washing out particulates from the troposphere. Similarly, as regarding the scavenging coefficient Λ, the effectiveness of particulate matter removal was dependent on the intensity and duration of the phenomenon of wet deposition. The comparison of areas that differ in terms of air quality indicated that the value of ΔC was affected by the location in which the process occurred. Concurrently, the season in the assessment of the method by means of which the concentration of particulate matter and intensity of precipitation were assessed, it was possible to assume that the variability of the results was affected by the aerosanitary condition specific for a given area. This situation was determined by the intensity of the local emission, scale of atmospheric dispersion and turbulence in the boundary layer of the troposphere. Despite the existence of a large degree of correlation between the duration and intensity of precipitation and the effectiveness of PM_{10} removal, it was difficult to develop a simple regression model that could be applied to the entire range of large-scale precipitation. The effect of other parameters besides intensity and duration of wet deposition was too large to secure the representative character of the presented models on a global scale. This also offered a justification for the considerable discrepancies between the results reported by various authors who conducted research in various climate zones. The special characteristics of wet deposition, as well as the location in which it occurred, impose a need to apply an individual approach to studying these problems. The instantaneous effect of purification as a result of the occurrence of wet deposition is minimised in the areas that are characterised by low air quality. Nevertheless, precipitation with considerable intensity leads to the extension of the period in which high environmental qualities are preserved.

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